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Absolute evaluation of magnetic ordering in correlated *f*-electron systems

Q. G. Sheng and B. R. Cooper

Department of Physics, West Virginia University, Morgantown, West Virginia 26506

J. M. Wills

Los Alamos National Laboratory, Los Alamos, New Mexico 87545

N. Kioussis

Department of Physics, California State University, Northridge, California 91330

For partially delocalized correlated *f*-electron systems, the key aspect of the electronic behavior is the hybridization of *f* electrons with the non-*f*-band electrons. This gives unusual properties including suppressed crystal-field splitting and highly anisotropic ordered magnetism. To improve the general understanding and to make the theory materially predictive, a technique is being developed to evaluate absolutely the parameters of the correlated electron model Hamiltonian, and then to use these to predict observed phenomenology including details of magnetic ordering such as magnetic structures and transitions between structures. The most difficult quantity to predict is the magnetic ordering temperature, among other reasons because it depends on the hybridization strength in a highly nonlinear way. Previously Wills and Cooper have reported on a technique involving a nonconventional electronic structure calculation based on treating the *f* electron as a resonant state in a solid-state environment to evaluate the hybridization. As an independent check on the evaluation of hybridization, here a conventional tight binding parametrization scheme was used to evaluate the hybridization. These results are compared both with previous results and with experiment for the magnetic ordering temperature and crystal-field dressing of Ce compounds, and the situation introduced by the relative degrees of nonlinearity for the crystal-field and magnetic ordering response to the hybridization strength are commented on.

Partially delocalized *f*-electron systems can be well described by the Anderson lattice Hamiltonian:

$$H = H_0 + H_1,$$

where

$$H_0 = \sum_k \epsilon_k n_k + \sum_{mR} E_m n_m(R) + U \sum_R N(R) [N(R) - 1]/2 \quad (1)$$

and

$$H_1 = \sum_{kmR} [V_{km}(R) c_k^\dagger c_m(R) + \text{h.c.}],$$

where ϵ_k and E_m are the band and *f*-state energies, U the intra-atomic *f*-*f* correlation energy, R labels the lattice site, and V_{km} is the hybridization matrix between the band and the *f*-state.

To make the theory able to predict the properties of a specific material on an absolute basis, and to improve our understanding, we have been developing techniques to evaluate from first principles the parameters in the Anderson Hamiltonian, especially the hybridization matrix V_{km} which plays the central role in the behavior of *f*-electron systems.¹

Once V_{km} has been evaluated, one can construct the function $V(\epsilon)$:

$$|V_M(\epsilon)|^2 = \sum_k V_{kM} V_{kM}^* \delta(\epsilon - \epsilon_k) / \sum_k \delta(\epsilon - \epsilon_k). \quad (2)$$

This quantity characterizes the strength and distribution of

the hybridization in energy space. If one then takes the product with the density of states $N(\epsilon)$, which characterizes the number of states available for hybridization, the quantity $\Gamma_M(\epsilon) \equiv \pi |V_M(\epsilon)|^2 N(\epsilon)$ (or its average over energy) characterizes the width of the localized *f* state due to its hybridization with the band states. As we will discuss below, experimental properties caused by hybridization can be evaluated by integrating $|V(\epsilon)|^2 N(\epsilon)$, or its products, as divided by various energy denominators.

In the work discussed here, we have evaluated the hybridization by using a tight binding parametrization scheme. On the basis of that evaluation, we have calculated the crystal-field dressing by the hybridization, and the two-ion interaction mediated by the hybridization.

Our calculation is for Ce mononictides, specifically on CeBi. Extrapolation from the heavier rare-earth mononictides² predicts crystal-field splitting that varies from 247 K for CeBi to 390 K for CeP. By contrast, the experimentally observed splitting varies from 8 K for CeBi to 150 K for CeP. An even more striking phenomenon observed in these systems is the strongly anisotropic magnetic ordering, with various phases having peculiar magnetic structures, and a Néel temperature of 25 and 16 K for CeBi and CeSb, respectively.³ Both the anomalous crystal-field splitting and anisotropic magnetic structure in Ce mononictides are believed to be the effect of band-*f* hybridization. Based on the Schrieffer-Wolff transformation,⁴ Wills and Cooper derived the expressions for the crystal-field level shift and the indirect *f*-*f* interaction.¹ We will use these expressions in evaluating these experimental properties.

The procedure for evaluating the hybridization is as follows:

(i) First we perform a warped-muffin-tin linearized muffin-tin orbital (LMTO) band structure calculation¹ for CeBi. The spin-orbit interaction is included perturbatively in the calculation. The potential within each muffin-tin sphere is approximated by a spherical average, while the potential in the interstitial region is expanded in a Fourier series. In Fig. 1, we show the LMTO density of states of CeBi calculated treating the $4f$ state as core; for comparison we have treated the $4f$'s as band states. The band states located in the vicinity of the Fermi level, which are therefore most physically relevant, are Ce- $5d$ and Bi- $6p$ bands. The position of the f level is found to be about 0.4 eV below the Fermi level. We also have varied this position, treating it as an adjustable parameter to study the effects.

(ii) Then we perform a Slater-Koster linear combination of atomic orbitals (LCAO)^{5,6} parametrization of the band structure. In this procedure, the p - p , d - d , and p - d interactions are considered with the first and the second neighbor, the f - p and f - d interactions are considered with the first neighbor, and the direct f - f interaction is neglected. We fit those eigenvalues for the region within 200 mRy above and below the Fermi level at 89 k-points of the irreducible Brillouin zone, with a rms deviation of 9.3 mRy. This compares, for example, with an rms deviation of 8.5 mRy in the fitting performed by Monnier, Degiorgi, and Koelling⁷ for YbP. In Fig. 2, we show the LCAO density of states from this fitting, and can compare with the LMTO density of states in Fig. 1. While quantitative details differ, the location of the peaked behavior with respect to the Fermi energy and the energy distribution of states giving the character of charge distribution (Bi- p , Ce- d) are quite similar; so that the fitting indicated by Fig. 2 is suitable for an alternative evaluation of hybridization. (One should bear in mind that the pertinent matrix elements connect states across the Fermi energy from one another.) The interactions between the f state and the non- f states are $(p\sigma) = 0.016$ Ry, $(p\pi) = -0.012$ Ry, $(d\sigma) = -0.004$ Ry, $(d\pi) = 0.002$ Ry, and $(df\phi)$ is small.

FIG. 1. The LMTO density of states of CeBi. The solid curve is the total density of states, the dashed curve and the dotted curve are the projected density of states for the Bi- p and Ce- d state, respectively. The Fermi energy is indicated by the vertical line.

FIG. 2. The density of states of CeBi obtained from LCAO parametrization fitting, with the various curves having the same meaning as in Fig. 1.

(iii) The LCAO fitting allows us to separate the electronic states into an assembly of bandlike states and an assembly of corelike states, while at the same time finding the matrix elements between them. At this point in the calculation, we have in hand a band state assembly composed of Ce- d and Bi- p states, the density of states of the bands, and the matrix elements between the band states and f states V_{km} . By projecting V_{km} into energy space using the tetrahedral scheme,⁸ we construct the function $|V(\epsilon)|^2$ defined in Eq. (2). For $j = \frac{5}{2}$ and cubic symmetry, V_M has two independent components: V_{Γ_7} and V_{Γ_8} , where Γ_7 and Γ_8 indicate the symmetry types. In Fig. 3 we show $|V(\epsilon)|^2$ for Γ_7 and Γ_8 states. One can see that $|V(\epsilon)|^2$ has peaklike behavior with its central region overlapping the position of the f level.

From the areas contained by the two curves, one can estimate that the Γ_8 state has stronger hybridization than the Γ_7 state roughly by a factor of 2-3. This will lead to a larger crystal-field shift for Γ_8 than for Γ_7 .

With the band structure and the hybridization thus obtained, we use expression (4.3) and (4.6) of Ref. 1 to calculate the crystal-field level shifts and the two-ion exchange

FIG. 3. The $|V(\epsilon)|^2$ function for CeBi. The dotted curve and the dashed curve are for the Γ_7 and Γ_8 state, respectively. The Fermi energy is indicated by the vertical line.

interaction (i.e., range parameters giving the interaction matrix between two neighboring f moments), respectively. The intra-atomic correlation U in these expressions is set to be infinite, and the f level in these expressions is placed 0.4 eV below the Fermi level.

We find that the Γ_7 and Γ_8 crystal-field levels shift downward by 51 and 160 K, respectively. This reduces the crystal-field splitting by 109 K. The crystal-field splitting suppression indicated experimentally is 239 K.¹ The crystal-field splitting suppression calculated using the resonance width scheme of Wills and Cooper is 217 K. For the range parameter, we find $E_1 = -0.28$ K, $E_2/E_1 = 2.0$, and $E_3/E_1 = -0.009$. Here E_1 , E_2 , and E_3 denote the dominant matrix component of the range parameter $E(1/2, 1/2)$ of the first, second, and third neighbor, respectively, where 1/2 indicates the quantum number along the interionic axis. Kioussis and Cooper expressed the temperature-dependent magnetic susceptibility in terms of the range parameter.⁹ At a certain temperature, the susceptibility has a singularity which implies a self-polarized magnetic ordering. By locating that singularity, we found the Néel temperature of CeBi to be 0.65 K. The experimental Néel temperature is about 25 times larger than the calculated value. The range parameters found by the resonance width scheme are $E_1 = 0.84$ K, $E_2/E_1 = 3.3$, and $E_3/E_1 = 0.08$, which also would imply a Néel temperature much below experiment. Thus our alternative scheme for evaluating hybridization gives results reasonably similar to Wills and Cooper¹ for observable quantities, considering the nonlinear relationship of those quantities to the hybridization. In both cases, the crystal-field dressing calculated is comparable with experiment, but the Néel temperatures are much smaller than the experimental value. The calculation of the Néel temperature is more difficult than that of the crystal-field suppression because of

its highly nonlinear dependence on the hybridization and f -state energy.

In addition to the hybridization-mediated interaction, there is another source of the two-ion interaction which is omitted from the present calculation, but may possibly have considerable contribution. That is the conventional RKKY interaction, which is additive to the hybridization-mediated interaction, as discussed by Cooper *et al.*³ If this increased the hybridization by a moderate factor, the Néel temperature would be increased corresponding to the fourth power of that hybridization; but the qualitative nature of magnetic behavior would be unchanged since it is determined by the highly anisotropic hybridization-mediated interaction. We are in the process of calculating this effect.

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